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Factors Influencing the Composition and Quantity of Passenger Car Refueling Emissions. Part 1

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by

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This paper describes the results of a study to examine the effects of various experimental variables on the quantity and composition of emissions associated with motor vehicle refueling. Problems related to accurate laboratory simulation of vehicle refueling are discussed. Preliminary results include emission rates for total hydrocarbons, benzene, toluene and 81 other hydrocarbon compounds for a single test vehicle.

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ABSTRACT

This paper describes the results of a study to examine the effects of various experimental variables on the quantity and composition of emissions associated with motor vehicle refueling. Problems related to accurate laboratory simulation of vehicle refueling are discussed. Preliminary results include emission rates for total hydrocarbons, benzene, toluene and 81 other hydrocarbon compounds for a single test vehicle.

RECENT CONCERNS ABOUT THE TOXICITY of benzene and adverse health effects associated with exposure to vapors from gasoline have renewed interest in vehicle refueling emissions. While refueling total hydrocarbon (THC) emissions data exists, little has been published about the emissions of specific hydrocarbon carcinogens. Therefore, this study focuses on the factors which influence emissions of benzene and other potentially carcinogenic hydrocarbon compounds during vehicle refueling.

Previous refueling studies have contributed significantly to an understanding of the primary mechanisms responsible for THC refueling losses and the variables upon which resultant emissions are dependent (1,2)*. These variables, which include fuel Reid vapor pressure (Rvp), dispensed fuel temperature (T_{DF}), vehicle tank fuel temperature (T_{VF}), and displaced vapor temperature (T_v), have been used to develop empirical models or equations to predict refueling losses. Use of the models is limited to the range of experimental conditions (i.e., fuel and vapor temperatures and fuel Rvps) investigated in these studies. Much of the information and insight gained in earlier studies has been applied here. Because the emission rates for

*Numbers in parentheses designate references at end of paper.

specific hydrocarbons are closely related to that for total hydrocarbons, the key variables cited above play an important role in this study. Effects resulting from varying the temperature of the delivery fuel and the vehicle fuel tank are examined. Fuel Rvp was not varied in this study; this parameter will be studied in a follow-on program which will examine seasonal variations.

In order to closely simulate refueling emissions of motor vehicles, complete motor vehicles with functioning evaporative emissions control systems were tested. Vehicle refueling took place in a specially equipped evaporative emissions measurement enclosure (SHED) with procedures similar to those of the SAE Recommended Practice (3). Hydrocarbon samples taken from the enclosure were analyzed with emission rates of THC, aromatics, olefins, paraffins, benzene, toluene, and 81 other HC compounds reported.

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EXPERIMENTAL PROCEDURES

Fuel Handling - The fuel handling scheme for the refueling experiments is shown in Figure 1. Gasoline stored in a 10,000 gal ground tank was first transferred to a 55-gal drum, then to a 60-gal refueling cart from which it was dispensed to the vehicle. When the refueling cart gasoline was not undergoing temperature conditioning, it was maintained at ambient temperature (72°F). Routine measurements of the Rvp and detailed hydrocarbon composition were found to be essentially the same as those for the ground tank gasoline. A comparison of hydrocarbon measurements depicted in Figure 2* indicates that no significant compositional changes occurred during fuel transfer or storage.

SHED Testing - The refueling experiments were conducted using a Sealed Housing for Evaporative Determination (SHED) technique (Fig. 1). The sealed housing or "SHED" was fitted with sampling ports, an air-mixing system, and a gasoline delivery hose and nozzle connected through gas-tight bulkhead fittings to the refueling cart. Prior to refueling tests, the SHED was examined for leaks and results indicated that it was in compliance with specifications set forth in the Federal Register (4) for air-tightness.

Prior to testing, the entire vehicle was placed within the SHED, the temperature of delivered fuel was stabilized at the test temperature, and background HCs were measured using a flame ionization detector (FID). The SHED door was then closed, and a person within the SHED uncapped the fuel tank and began fueling the vehicle. Fuel flow was normally maintained at about 8 gallons per minute (gpm) through a standard unleaded gasoline dispensing nozzle. When the vehicle tank was full or had been refueled as necessary, the nozzle was shutoff, and the fuel tank was capped. Hydrocarbons in the SHED were continuously monitored with an FID to determine when the vapor had become

* All hydrocarbon data in this study are based on carbon concentration, not molar concentration

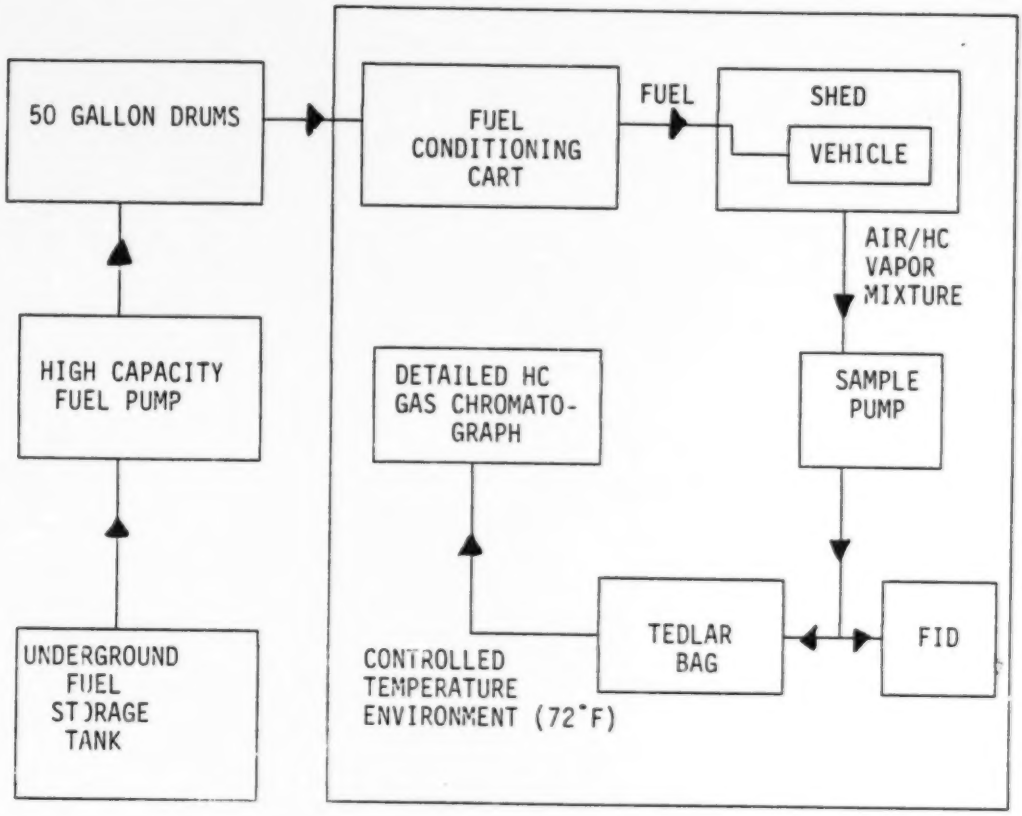

Fig. 1 - Fuel Handling and Experimental Setup

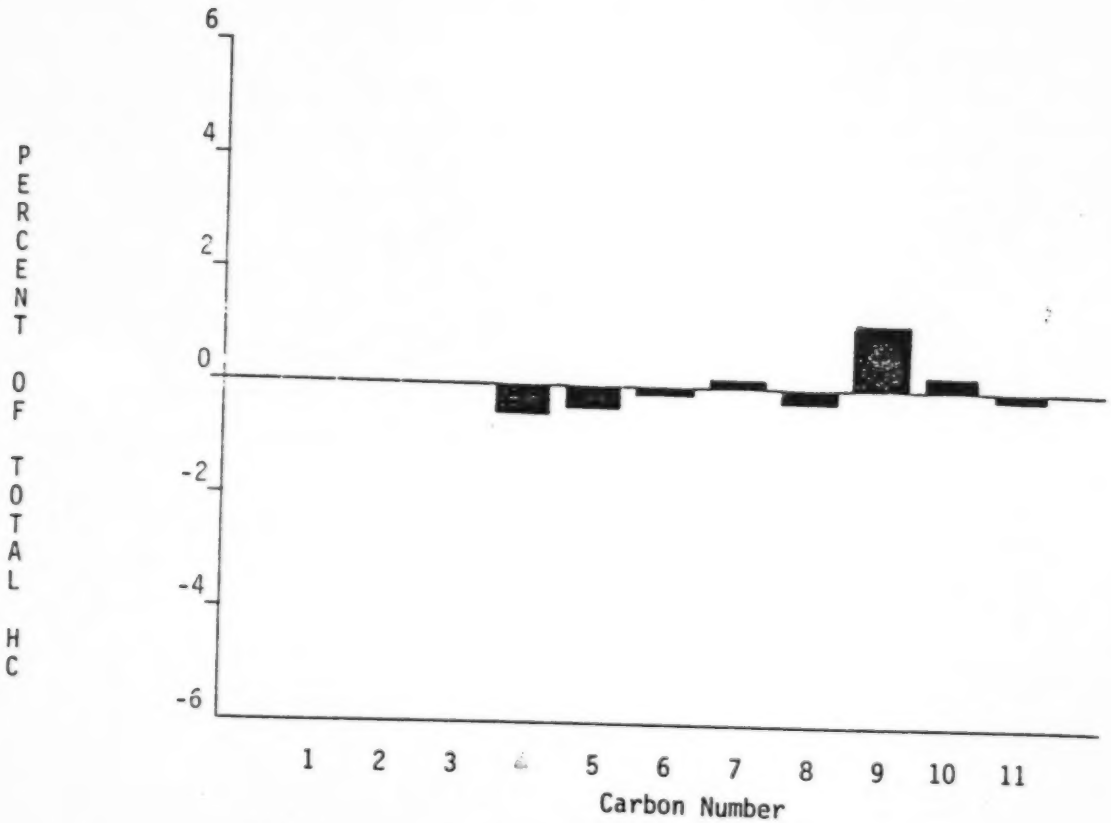

Fig. 2 - Difference of detailed HC analyses
10K gallon tank minus refueling cart.
Hydrocarbons grouped by carbon number.

thoroughly mixed with the SHED air. Normally, HC levels stabilized about five minutes after replacement of the gas cap. At that time the FID was calibrated with known concentrations of propane in air, and a total HC measurement was taken. In addition, a SHED sample was collected in a 5-L Tedlar bag for subsequent gas chromatographic (GC) analysis. The GC was programmed to identify 83 different C_1 - C_{11} HCs. The detailed HC analyses were performed using procedures previously published (5). Temperature and pressures in the SHED and temperatures of the delivered fuel (T_{DF}) and the vehicle's tank fuel (T_{VF}) were recorded at the beginning of the test. The vehicle's tank contained 2 gal of fuel before refueling providing vapor to the tank's head space.

Of utmost concern in this test procedure were the hazards associated with handling gasoline within confined spaces. The individual delivering the fuel was equipped with SCUBA gear to minimize the likelihood of breathing harmful vapors. Care was also taken to prevent spills or other situations that might result in a fire or explosion hazard. Fire extingushers were in place and personnel were trained in procedures to follow in the event of a gasoline spill. Although HC concentrations in the SHED remained well below 15,000 ppm, precautions were in place to purge the SHED enclo re had concentrations exceeded that level. This allowed an adequate margin for safety since the lean flammability limit for gasoline is 60,000 ppm.

HC MASS MEASUREMENT - The mass emission rate of HC from the refueling operation was calculated using procedures outlined in the Federal Register for evaporative emissions determination (4). An alternate procedure involving the vaporization of known quantities of gasoline inside the SHED to develop a relationship between HC concentration and mass was also used. Comparison of these methods agreed within 10%.

RESULTS AND DISCUSSION

This study was conducted to examine the effect of refueling variables on the quantity and composition of refueling emissions. Many of the variables have already been studied, often with simulations that were unlike the full-scale or complete vehicle simulations used in this study. Therefore, further study of variables such as fuel delivery rate, fuel tank capacity and shape, fill tube length and configuration, gasoline volatility, tank and delivery fuel temperatures, was needed to determine their importance to emission rate and composition.

Fuel delivery rate - Gasoline having a Rvp of 11.4 psi and a temperature of 59°F was delivered to a 1978 Ford Mustang at three flow rates (Q_D). Approximately 13 gal were dispensed into a tank initially containing 2 gal of gasoline at 73°F. Emission results for the three flow rates are shown in Table 1. Fuel delivery rates ranging from 2 to 8 gpm did not significantly effect g/gal THC emission rates, but could influence instantaneous exposure because the emission rate in g/min would vary with fuel delivery rate.

Fuel tank capacity and shape - Tests were performed on three vehicles having different fuel tank capacities and configurations. Fill tube lengths and shapes were also different in each case. Results presented in Table 2 support the contention that shape and size variables had no significant effect on g/gal THC emission rates. Under the conditions of these tests (i.e., T_{VF} = 73°F, T_{DF} = 59°F, Rvp = 11.4 psi, and Q_D = 8 gpm), emissions were

Table 1. Effects of Fuel Delivery Rate on Total HC Fueling Emissions Rate

Fuel Delivery Rate (gal/min)	Total HC Emissions (g/gal)
2.0	4.27
4.6	4.61
8.0	4.61

Table 2. Effects of Vehicle Type and Configuration on Total HC Fueling Emissions Rate

Model Make	1976 Ford Mustang	1984 Dodge RAM Pickup	1985 Chev. Caprice
Fill Tube Location	L,R,Q,P*	L,R,Q,P	Rear, Center
Fuel Tank Cap. (Gal)	15	20	25
Fuel Delivered (Gal.)	13	18	23
Total HC Emissions (gm/gal)			
Mean (3 Tests)	4.54	4.55	4.51
Std Dev.	0.34	0.10	0.28

* LRQP = Left Rear Quarter Panel

observed to be linearly proportional to the amount of fuel transferred.

Source of emitted HC vapor - Several tests were conducted to determine whether the HC vapors emitted during refueling were primarily from the fuel tank head-space vapor or from the dispensed fuel volatiles. In one test a 1978 Ford Mustang initially containing 2 gal with an Rvp of 11.4 psi gasoline was fueled with 13 gal of water at 55°F. THC mass emissions were 3.95 g/gal, which is 87% of the THC average value obtained when the same vehicle was fueled under similar conditions with gasoline (Table 3). These results are consistent with findings by Smith (1), who conducted bottom and top-fill experiments to isolate emissions caused by entrained droplet loss during top-fill refueling. Only a 10% increase in refueling emissions was observed when the tank was top-filled.

The vapors emitted in the water-fill test were also subjected to detailed HC analysis with a gas chromatograph. Figure 3 shows a comparison of carbon number distributions for vapors from the gasoline and water-filled tests. Figure 3 depicts the difference, carbon number by carbon number, between the water-fill test and a gasoline-fill test made under similar conditions. It shows that the gasoline-fill test vapors have relatively greater concentrations of the lighter hydrocarbons (C_4) and indicates that the contribution of the dispensed gasoline stream to the refueling emissions composition is primarily lighter hydrocarbons.

Temperature effects - A series of tests were conducted to investigate the effects of varying the temperatures of both the vehicle fuel tank and the dispensed fuel. In these tests 2 gal of cold fuel was placed in the tank of a 1978 Ford Mustang (15 gal capacity) and brought to the desired temperature using a heat blanket. Rvp of the fuel was 11.4 psi. When the desired tank and dispensed fuel temperatures were attained, fuel was dispensed to the

Table 3. Comparison of Total HC Mass Emission Rates for Water Fill and Gasoline Fill Tests

	T_{DF} (°F)	T_{VF}(°F)	Total Fill (gal)	Total HC (gm/gal
Water Fill	55	72	13	3.95
Gasoline Fill	59	72	13	4.54

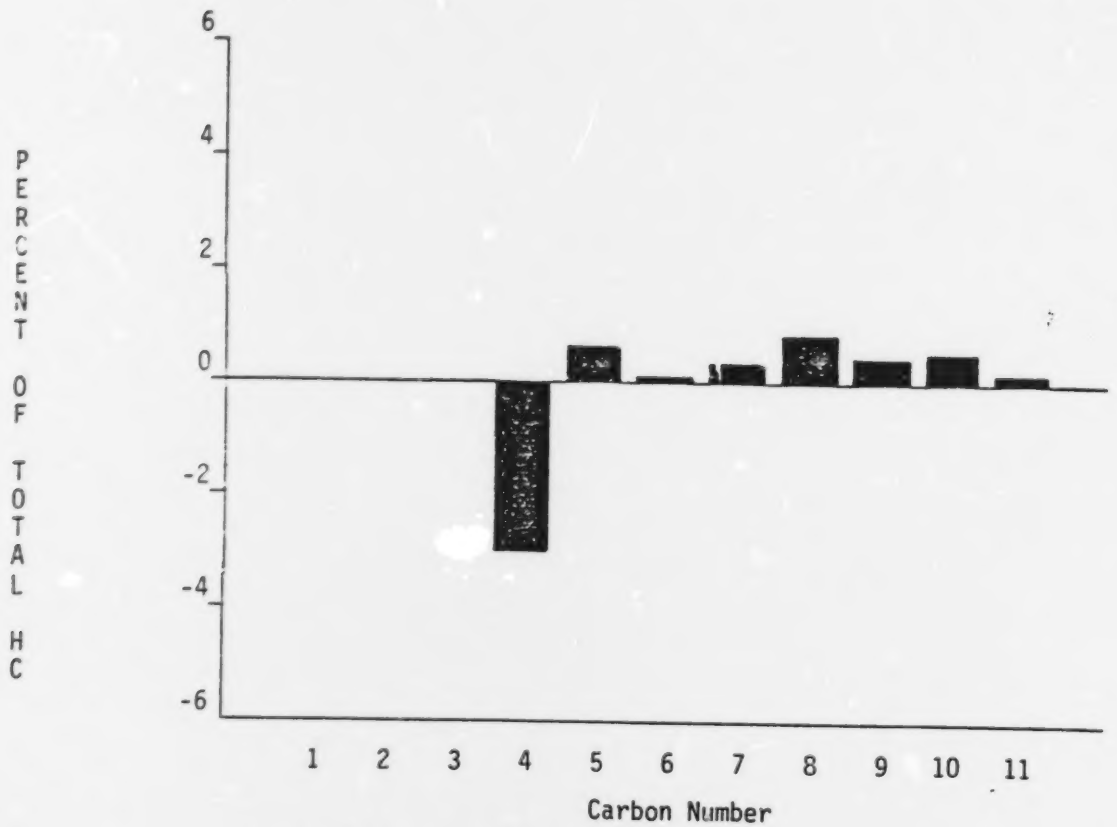

Fig. 3 - Difference in Refueling Vapor Composition
Water Fill Minus Gasoline Fill
Hydrocarbons Grouped by Carbon Number.

vehicle.

Figures 4, 5, and 6 depict mass emissions (normalized to the value obtained at T_{DF} = 59°F, T_{VF} = 85°F) measured for various combinations of T_{DF} and T_{VF} for total hydrocarbons, benzene, and toluene. Normalization to temperatures more representative of summertime conditions is appropriate since a summer commercial fuel was being used. In general, emissions appear to be sensitive to both fuel tank temperature and dispensed fuel temperature. The magnitude of vapor growth or shrinkage in a fuel tank is dependent on the difference between T_{VF} and T_{DF}. Shrinkage and associated emissions decreases occur when T_{DF} is less than T_{VF} (common during summer months). Vapor growth occurs when T_{DF} is greater than T_{VF} (common during winter months).

Fuel aging tests - Refueling emissions have been observed to increase slightly when aged fuel rather than fresh fuel is used in the vehicle tank (6). A prime concern in the laboratory simulation was that the heat blanket method used to elevate the vehicle's fuel tank temperature would age or weather the fuel in an unrealistic manner. For example, if the gasoline light-ends were being boiled off to a greater extent in the heat blanket simulation, the vapor pressure of the tank fuel would be artificially depressed. To explore this possibility, three tests were conducted to evaluate fuel aging during over-the-road operation of the vehicle.

In two of the tests, a 1978 Ford Mustang was refueled with 5 gal of gasoline and driven for 45 min over a road course consisting of about 90% interstate highway. The temperature was 85°F and skies were sunny for both of these tests. The third test was conducted similarly except that the vehicle's fuel tank was completely filled, and the vehicle was driven on the road for about 30 miles per day for a seven-day period. The vehicle was parked outside in weather that was mostly sunny with high temperatures averaging around 85°F

T_{VF} (°F)	92	76	59	45
120	0.65	0.89		
85	2.14	1.51	1.00	
50		2.02	1.54	1.2
15			1.80	1.31

T_{DF} (°F)

Fig. 4 - Normalized Total HC for Various T_{DF} and T_{VF}

T_{VF} (°F)	92	76	59	45
120	0.78	1.09		
85	2.44	1.56	1.00	
50		2.17	1.56	1.17
15			1.86	1.39

T_{DF} (°F)

Fig. 5 - Normalized Benzene for Various T_{DF} and T_{VF}

T_{VF} (°F)	92	76	59	45
120	0.85	1.57		
85	2.28	1.57	1.00	
50		2.00	1.42	1.14
15			1.71	1.29

T_{DF} (F)

Fig. 6 - Normalized Toluene for Various T_{df} and T_{VF}

and lows around 65°F.

After each road run was completed, the vehicle was returned to the laboratory where refueling tests were conducted. In addition to THC refueling emissions, measurements taken before and after the road runs included vehicle fuel tank gasoline RVP and carbon number distribution, carbon number distributions of the HC refueling emissions, T_{VF}, and T_{DF}. Results are presented in Table 4 and Figures 7 through 12.

In Table 4, the values of refueling vapor THC mass emissions have again been normalized to the value at T_{DF} = 59°F, $T_{V}F$ = 85°F of Figure 4. This permits a comparison with data previously cited in Figure 4 where the vehicle fuel tank was heated by a thermal blanket. The normalized THC mass emission rate of 0.97 given in Table 4 for T_{DF} = 76°F, T_{VF} = 106°F is slightly higher than that of 0.89 given in Figure 4 for T_{DF} = 76°F, T_{VF} = 120°F. This is consistent with the trend observed when using the heat blanket wherein an inverse relationship existed between mass emissions and T_{VF}. Furthermore, examination of Table 4 shows that for each of the three road tests the fuel in the vehicle tank decreased in Rvp and increased in average carbon number. This suggests that fuel aging had occurred. As expected, the vapor emitted during the final fueling increased in average carbon number from that initially measured due to the loss of light HCs in fuel. Shifts in carbon number in the fuel and vapor are illustrated in Figures 7 - 12. These show the before and after differences between carbon number distribution analyses of vehicle fuel tank gasoline and fueling vapor for each of the three road tests.

Figures 11 and 12, which show the data for the seven day test, are not very different from the Figures for the 1 hour tests, indicating that a large

Table 4. Results of Fuel Aging Tests

	Test 1 (45 min)	Test 2 (45 min)	Test 3 (7 day)
Normalized Refueling Vapor Emissions Rate	0.64	0.97	0.92
Vehicle Tank Temp (°F)	106	102	106
Vehicle Tank Fuel RVP			
Initial	10.6	10.7	11.0
Final	9.9	10.2	9.6
Vehicle Tank Fuel Avg. Carbon No.			
Initial	7.52	7.46	7.48
Final	7.65	7.71	7.64
Fueling Vapor Avg. Carbon No.			
Initial	4.97	4.99	4.94
Final	5.21	5.17	5.20

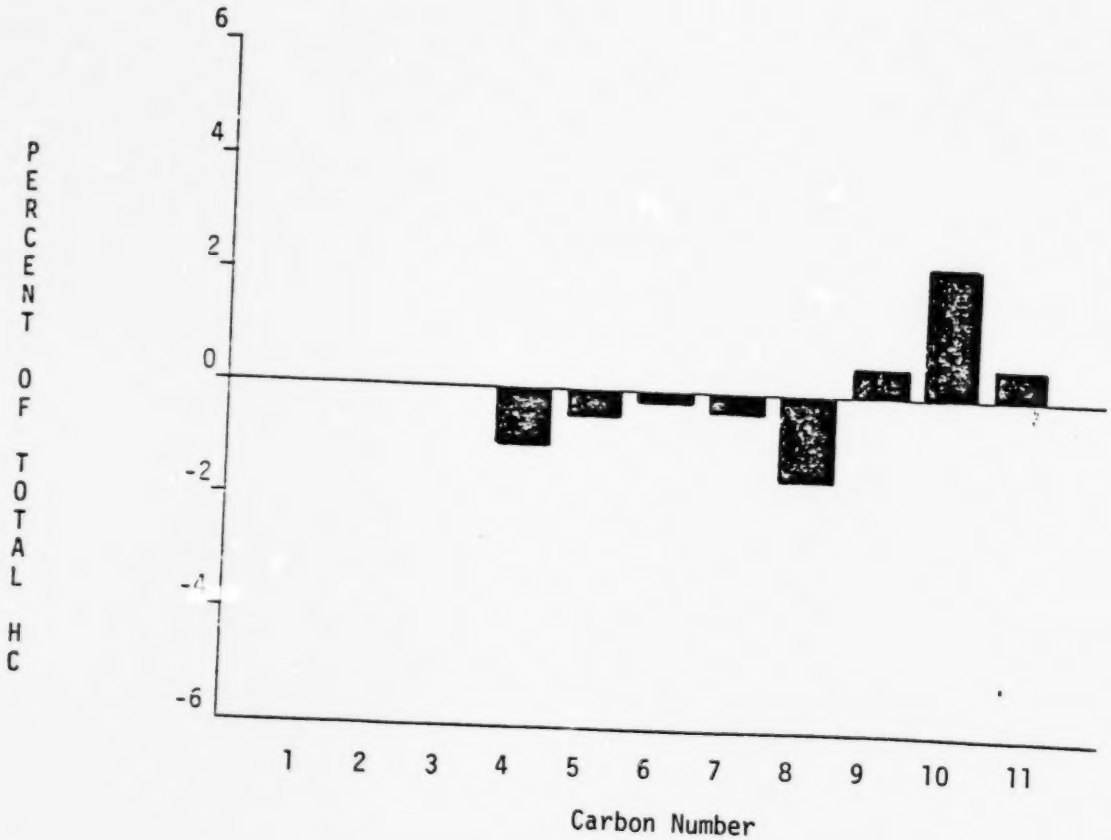

Fig. 7 – Difference in Gasoline Composition Post-Aging Minus Pre-Aging (1 Hr Test). Hydrocarbons Grouped by Carbon Number.

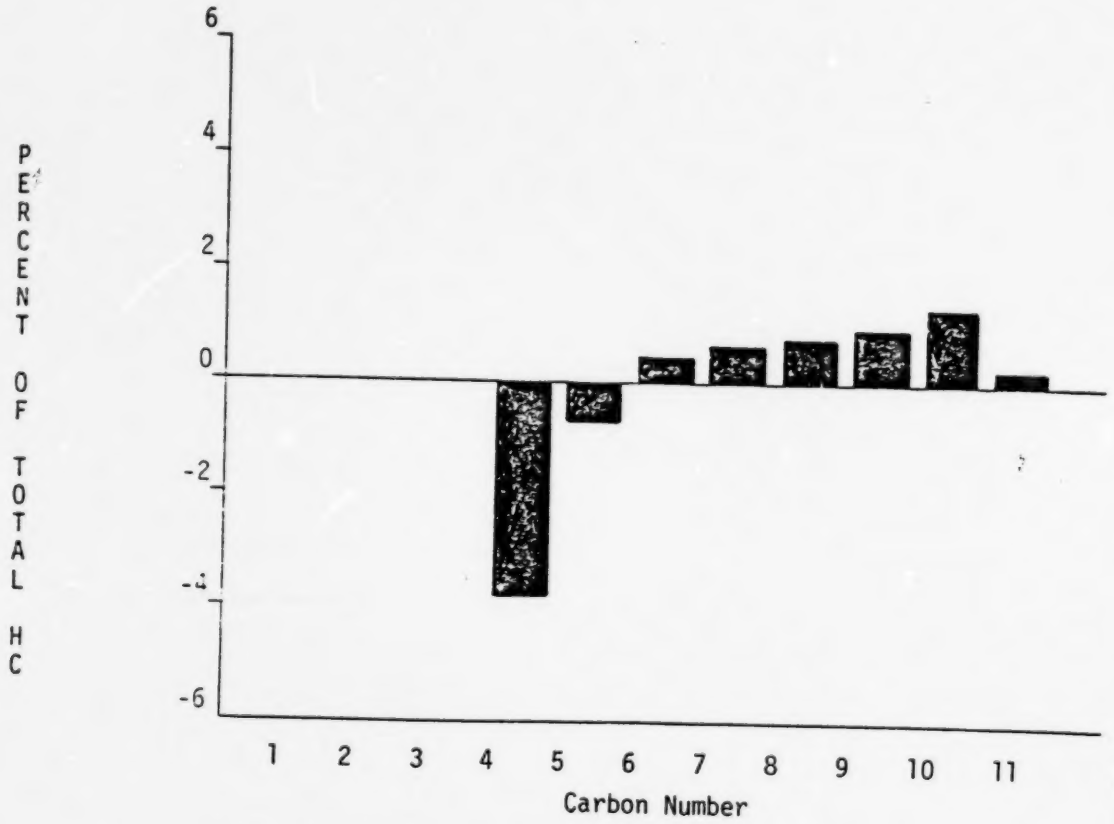

Fig. 8 - Difference in Refueling Vapor Composition Post-Aging Minus Pre-Aging (1 Hr Test). Hydrocarbons Grouped by Carbon Number.

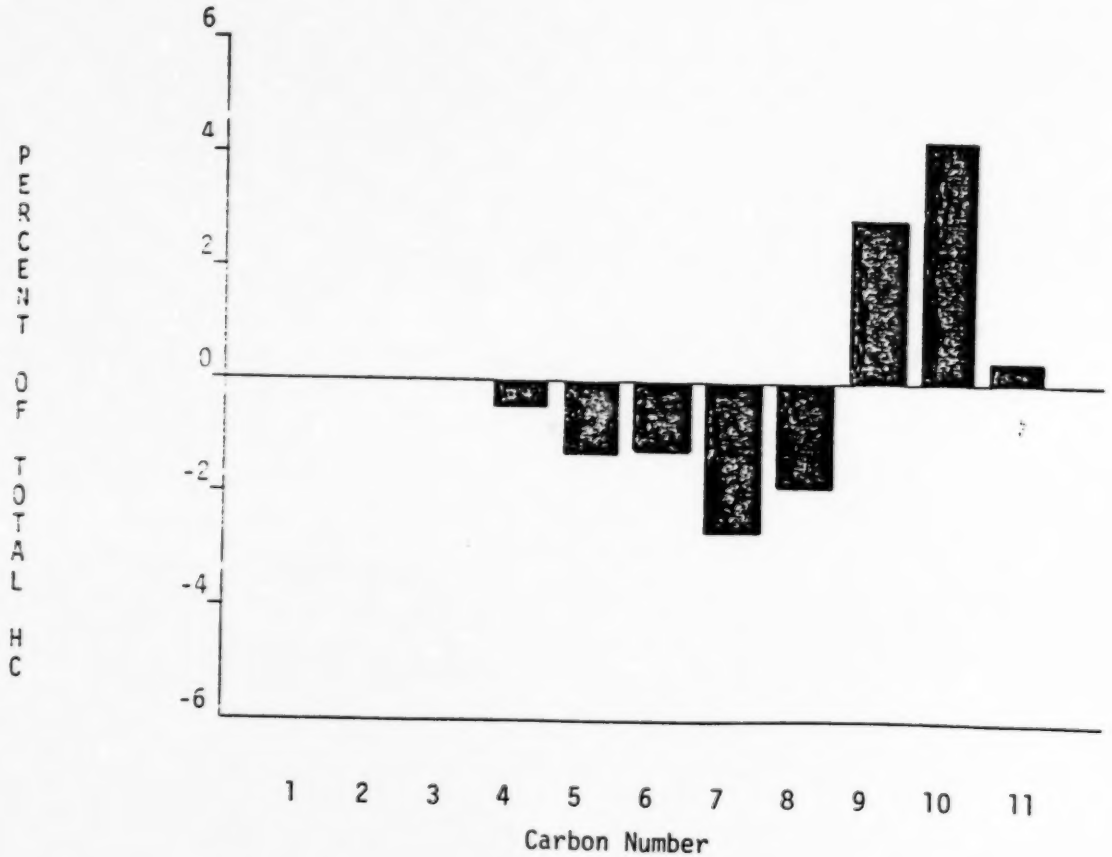

Fig. 9 - Difference in Gasoline Composition Post-Aging Minus Pre-Aging (1 Hr Test). Hydrocarbons Grouped by Carbon Number.

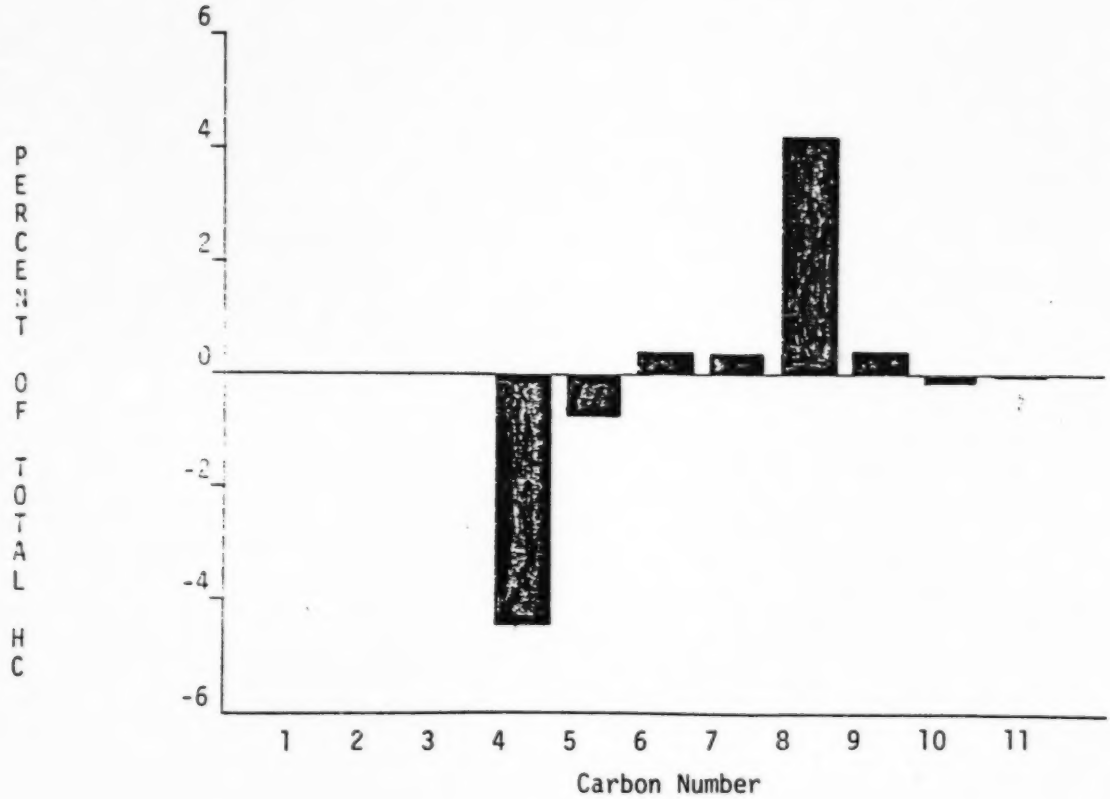

Fig. 10 - Difference in Refueling Vapor Post-Aging Minus Pre-Aging (1 Hr Test). Hydrocarbons Grouped by Carbon Number.

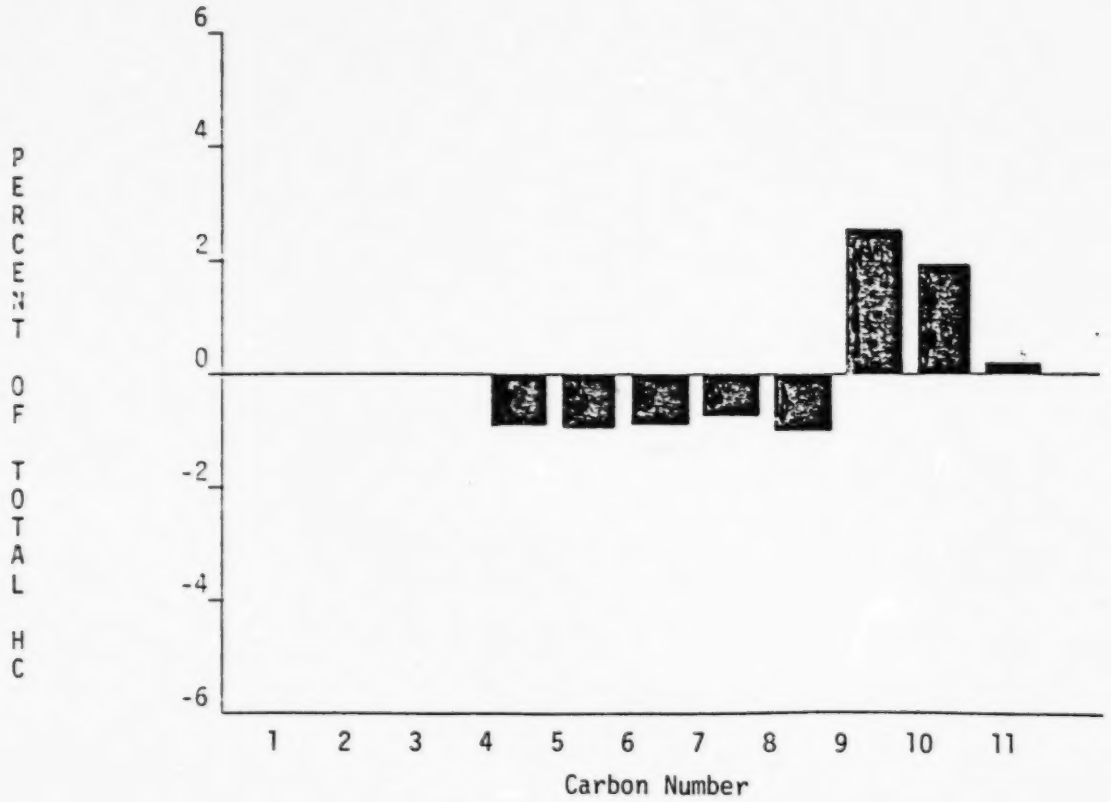

Fig. 11– Difference in Gasoline Composition Post-Aging Minus Pre-Aging (7 Day Test). Hydrocarbons Grouped by Carbon Number.

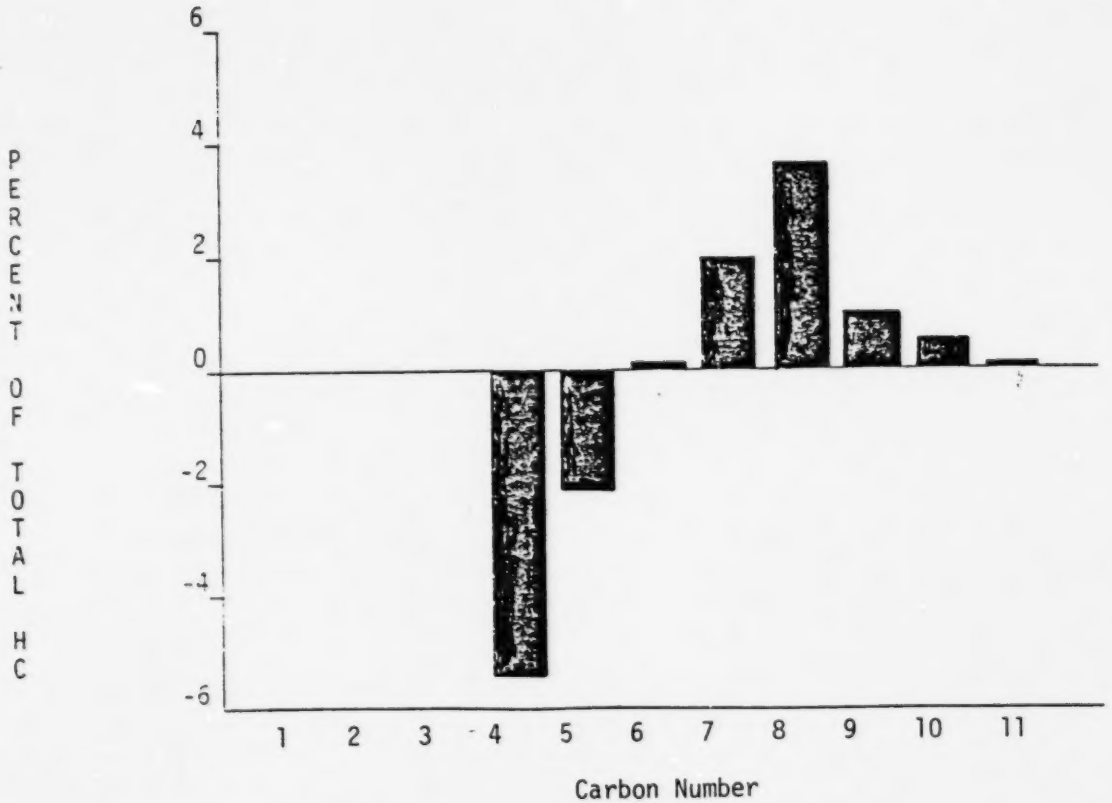

Fig. 12 – Difference in Refueling Vapor Composition Post-Aging Minus Pre-Aging (7 Day Test). Hydrocarbons Grouped by Carbon Number.

part of this compositional change occurred during the first hour of over-the-road operation after fueling. It appears that fuel aging could influence the composition of refueling emissions and therefore, will be further studied in the follow-on program to examine seasonal variaions.

Appendix A presents detailed hydrocarbon results for tests conducted at T_{VF} = 72°F and T_{DF} = 59°F. Similar data will be developed in the follow-on seasonal study.

CONCLUSIONS AND RECOMMENDATIONS

The findings of this study confirm the conclusions of earlier work that vehicle fuel tank size, shape, configuration, and location; and size and length of the fill tube; and fuel delivery rate have no significant effect on the total mass of motor vehicle refueling emissions. The total mass of HC vapors emitted during vehicle fueling is directly proportional to the amount of fuel delivered to the tank. The emissions result primarily from displacement of the vapors in the vehicle tank headspace.

Fueling vapor emissions are also dependent on the temperature of both the delivered fuel and the fuel in the vehicle tank. In this study emissions generally decreased with a decrease in delivered fuel temperature and an increase in vehicle tank fuel temperature. Findings of a brief examination of fuel aging suggest that emissions composition may be influenced by aging and that further study is needed to assure that the heat-blanket method is suitable for laboratory simulation. In general, interrelationships between the composition of refueling emissions, tank fuel Rvp, and fuel aging require further definition. A follow-on program to be conducted by EPA will address these interrelationships while examining refueling losses for typical fuels used in winter, spring, and summer under normal use conditions.

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APPENDIX A. Detailed hydrocarbon characterization of refueling emissions for T_{VF} = 72°F, T_{DF} = 59°F

Figures A1 and A2 present hydrocarbon carbon number distributions of the test fuel and refueling emissions, respectively. Table A1 presents individual hydrocarbon emission rates for refueling at T_{VF} - 72°F, T_{DF} = 59°F.

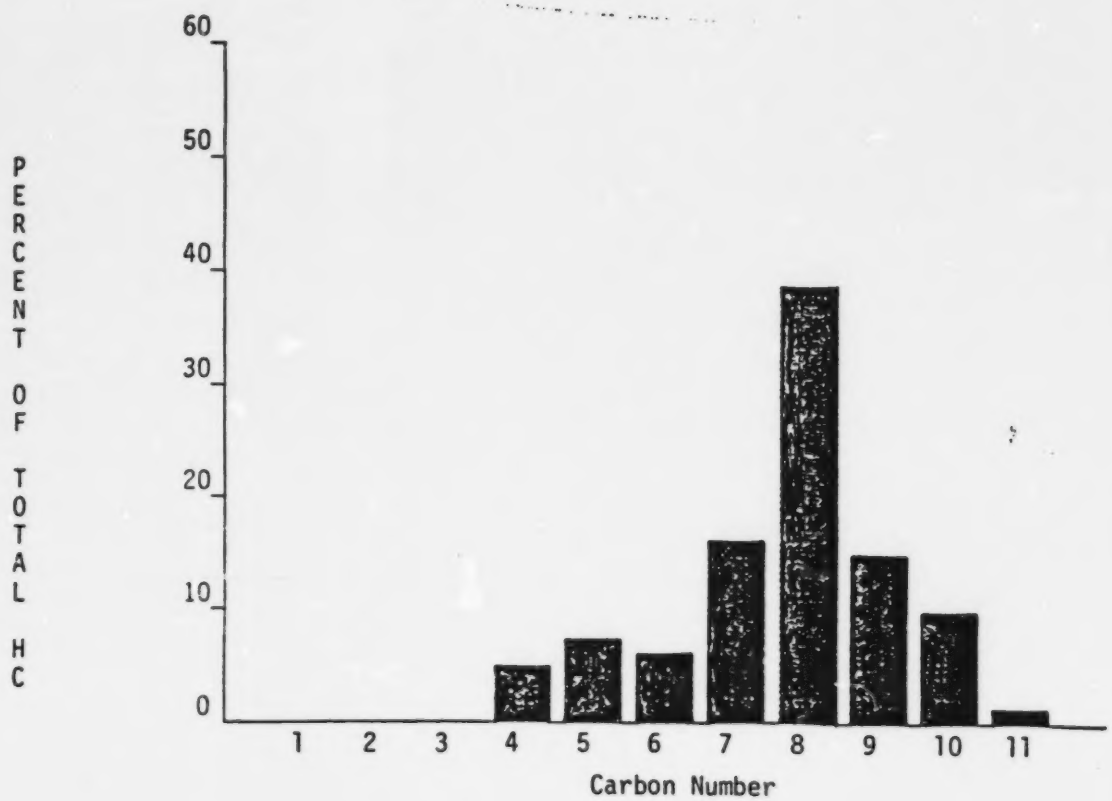

Fig. A1 Typical Refueling Cart Gasoline Composition. Hydrocarbons Grouped by Carbon Number.

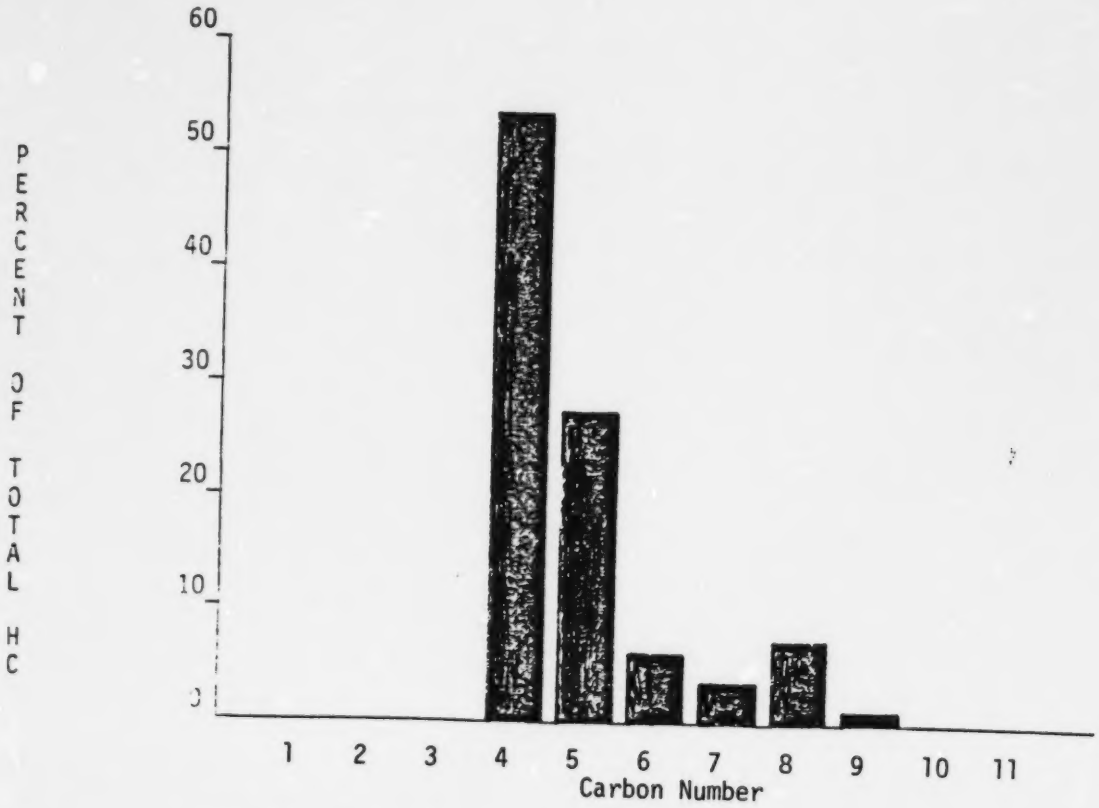

Fig. A2 Refueling Vapor Composition at $T_{VF} = 72^{o}F$, $T_{DF} = 59^{o}F$. Hydrocarbons Grouped by Carbon Number.

Table A1. Mass HC Emission Rates for 83 Compounds

T_{VF} = 72°F, T_{DF} = 59°F (g/gal of dispensed fuel)

Compound	Mass (g/gal)
1. Methane	0.0
2. Ethylene	0.0
3. Ethane	0.0
4. Acetylene	0.0
5. Propylene/Propane	0.0
6. Propadiene	0.0
7. Methylacetylene	0.0
8. Isobutane	0.275
9. Butene-1/Isobutylene	0.063
10. N-Butane/1,3-Butadiene	1.785
11. Trans-2-Butene/2,2-Dimethylpropane	0.075
12. Cis-2-Butene/1-Butyne	0.079
13. 3-Methyl-1-Butyne	0.018
14. Isopentane	0.777
15. Pentene-1/2-Butyne	0.035
16. N-Pentane	0.138
17. 2-Methyl-1,3-Butadiene	0.002
18. Trabs-2-Pentene	0.064
19. Cis-2-Pentene	0.030
20. 2-Methyl-2-Butene	0.064
21. 2,2-Dimethylbutane	0.006
22. Cyclopentene	0.010
23. Cyclopentane/3&4-Methyl-1-Pentene	0.010
24. 2,3-Dimethylbutane	0.047
25. 2-Methylpentane/2,3-dimethil-1-butene	0.068
26. 3-Methylpentane	0.035
27. 1-Hexene/2-ethyl-1-butene	0.006
28. N-Hexane/Cis-3-Hexene	0.018
29. C6 Olefin	0.009
30. 2-Methyl-2-Pentene	0.023
31. 2-Hexene (Cis & Trans)	0.004
32. Methylcyclopentane/3-Methyl-Trans-2-Pentene	0.026
33. 2,4-Dimethylpentane	0.025
34. Methylcyclopentene	0.003
35. Benzene	0.029
36. Cyclohexene/2,3-Dimethylpentane/2-methylhexane	0.034
37. 3-Methylhexane	0.014
38. 2,2,4-Trimethylpentane	0.065
39. N-Heptane	0.007
40. Methylcyclohexane	0.004
41. Dimethylhexene	0.008
42. 2,2-Dimethylhexane	0.002
43. 2,4 & 2,5-Dimethylhexane	0.011
44. 2,3,4-Trimethylpentane	0.016
45. 2,3,3-Trimethylpentane	0.019
46. Toluene/2,3-Dimethylhexane	0.080
47. 2-Methylheptane/1-methylcyclohexene	0.004
48. 4-methylcheptane	0.003

49. 3-methylheptane	0.005
50. 2,2,5-Trimethylhexane	0.002
51. N-Octane	0.002
52. 2,3,5-Trimethylhexane	0
53. 2,4-Dimethylheptane	0.005
54. 2,5 & 3,5-Dimethylheptane	0
55. Ethylbenzene/2,3-Dimethylheptane	0.009
56. P&M - Xylene	0.022
57. 2-Methyloctane	0
58. 2,4,5-Trimethylheptane	0
59. O-Xylene/Unk C9 Paraffin	0.016
60. 2,4-Dimethyloctane	0.001
61. N-Nonane	0.001
62. Unknown (C10 Paraffin)	0
63. N-Prophylbenzene	0.008
64. 1,3,5-Trimethylbenzene	0.006
65. 3,4-Dimethyloctane	0.004
66. 1-Methyl-3-Ethylbenzene	0.003
67. 1-Methyl-2-Ethylbenzene	0
68. 1,2,4-Trimethylbenzene	0.013
69. Secbutylbenzene	0
70. N-Decane	0.004
71. 1,2,3-Trimethylbenzene	0
72. Indane	0.002
73. Isobutylbenzene	0.004
74. 1-Methyl-3N-Propylbenzene	0
75. 1,3-Diethylbenzene	0
76. 1-Methyl-3-Isopropylbenzene	0
77. 1,2-Diethylbenzene	0
78. 2-Methyldecane	0
79. C10 Aromatic	0.003
80. C10 Aromatic	0
81. N-Undecane	0
82. C10 Aromatic	0.001
83. Unknowns	0.123

Total HC Mass Emission Rate: 4.23 g/gal